Pressure dependence of the low-frequency dielectric constant of KNbO$_3$

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Abstract

The effect of pressure on the low-frequency dielectric constant, ϵ_0, of single crystals of KNbO$_3$ is investigated by means of capacitance measurements. The dielectric constant increases with pressure up to 22.5 kbar, where it exhibits a large value ($\epsilon_0 = 5000$), and then decreases. This change in its behaviour is related to a phase transition induced by pressure. On decompression, the samples do not revert back to the ambient pressure phase.

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Potassium niobate (KNbO$_3$), which is a prototype of a ferroelectric material, at ambient conditions presents an orthorhombic structure. Recently, KNbO$_3$ has been studied under hydrostatic pressure showing that this compound undergoes some structural changes[1, 2]. By Raman scattering several phase transitions have been reported[2]. The pressure-induced phases are different from those induced by temperature at ambient pressure. In particular, a displacive type phase transition has been proposed at 20 kbar. This transition is mainly characterized by softening of a transverse optical (TO) mode with wavelength $\omega_{TO} = 50$ cm^{-1}. However, its transition pressure, P_t, is not well established since the mode frequency cannot be detected above 20 kbar due to the disappearance of this mode into the continuous background. The interest in more detailed information about this phase transition stimulated the present measurements of the low-frequency dielectric, ϵ_0, constant under pressure up to 32 kbar, at room temperature.

The KNbO$_3$ crystal samples used in the experiments were grown from a melt comprised of a mixture of K$_2$CO$_3$ and NbO$_5$. High-resistivity single crystals were oriented along [010] direction, being ϵ_0 in this orientation larger than in the other axis. Samples for the capacitance measurements were cut and polished into slabs 100-200 μm thick and 2x2 mm^2 in size. Silver electrodes were vacuum evaporated on the large sample faces. Ohmic contacts were made by soldering silver wires to the electrodes with high-purity indium. The capacitance of the samples was measured by using a high-accuracy ($> 0.1\%$) capacitance meter and shielded leads. High-pressure was generated by a Bridgmann cell which has been described in earlier studies[3]. In this case, we have used tungsten carbide anvils, 27 mm in diameter, without steel binding rings and sodium chloride was the pressure-transmitting medium. The pyrophyllite gaskets employed, which were treated at 720 ^{o}C during one hour in order to get suitable mechanical properties, were 0.5 mm in thickness with a hole of 9 mm in diameter. The pressure was determined by calibration of the load applied to the anvils against known fixed points.

The capacitance, C, at a given pressure, P, is given by:

$$C(P) = \epsilon_0(P)\frac{A(P)}{d(P)} \quad ,$$

(1)

where A is the area of the silver electrodes and d is the slab thickness. Then, through

Eq. (1) we can obtain $\epsilon_0(P)$ from $C(P)$, provided one takes into account the changes in the sample dimensions due to the compression. For this purpose, we have used a Murnaghan equation of state with the bulk modulus B_0=1420 kbar and B_0'=4 as deduced by x-ray diffraction[4]. Figure 1 shows the results of ϵ_0 as a function of the applied pressure for a sample with $d = 100$ μm. There it can be seen that, under compression, ϵ_0 increases until it reaches its maximum value ($\epsilon_0 = 5000$) at about the pressure of 22.5 kbar. In particular, ϵ_0 increases nearly linear up to around 15 kbar, above this pressure the slope $d\epsilon_0/dP$ changes from 75 kbar^{-1} to 180 kbar^{-1}. Finally, above 22.5 kbar ϵ_0 decreases monotonically. We think, that the drastict change in the behaviour of the low-frequency dielectric constant at 22.5 kbar is originated by the phase transition induced by pressure reported in Ref. [2]. The roundness of the observed peak may be related to the quasi-hydrostatic conditions at which we carried out the experiments. Note that unavoidable uniaxial stresses and sample deformations, due to the solid pressure-transmitting medium used, can contribute to reinforce crystal instabilities. Fig. 1 also shows that on decompression ϵ_0 does not increase as expected for a reversible phase transition. This attests an apparently non-reversible phase transition, where the high-pressure phase remains stable at ambient conditions after releasing the pressure. The results have been tested for their reproducibility in five different samples. Then, by averaging the pressure where the maximum of ϵ_0 occurs we estimated that $P_t = 22.5 \pm 0.5$ kbar

The results obtained below P_t can be checked by calculating indirectly the Grüneisen parameter γ of the 50 cm^{-1} TO mode. In ferroelectric materials, as a first approximation, the relationship between ϵ_0 and the TO mode wavelength is given by the Lyddane-Sachs-Teller relation. Under this assumption, it is easily to show that the Grüneisen parameter of ω_{TO} is given by:

$$\gamma = \frac{-B_0}{2} \left(\frac{d \ln \epsilon_0}{dP} \right)_{P=0} . \qquad (2)$$

Then, from our results we get γ = -29. This large value of γ indicates that a small change in volume pruduces a relatively large change in ω_{TO} as was previously stablished[2].

In conclusion, our results confirm that a non-reversible phase transition induced by

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pressure occurs at around 20 kbar. In addition, we have estimated that $P_t = 22.5 \pm 0.5$ kbar.

Acknowledgments

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Figure captions

Figure 1: Pressure dependence of the static dielectric constant of $KNbO_3$. Symbols represent the data obtained during compression ($\circ$) and decompression ($\bullet$).